Mass transfer in gas sparged membrane cells

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Rates of mass transfer for the transfer of NaCl across a high permeability Cuprophane membrane in a gas-stirred batch dialysis cell were measured by the conductivity method. Gas sparging was found to increase the rate of mass transfer across the membrane by an amount ranging from 18 to 360% depending on the superficial gas velocity and the physical properties of the solution. The overall mass transfer coefficient was related to the superficial velocity by the equation

$$K = aV_{\rm s}^{0.22}$$

The implication of the results for industrial membrane separation processes such as dialysis and electrodialysis is discussed.

Nomenclature

| A | membrane area |
|---------------|---|
| a, a_1, a_2 | , a_3 and b constants |
| C_1 | salt concentration at time t in compartment 1 |
| C_2 | salt concentration at time t in compartment 2 |
| C_0 | initial salt concentration in compartment 1 |
| D | diffusivity of the salt |
| Fr | Froude number (V_s^2/lg) |
| g | acceleration due to gravity |
| Ι | cell current |
| J | mass transfer J factor ($St Sc^{0.66}$) |
| K | overall mass transfer coefficient |
| K_1, K_2 | liquid phase mass transfer coefficients |
| , | |

l membrane height

1. Introduction

In membrane cells used in dialysis, electrodialysis, hyperfiltration and reverse osmosis, the maximum rate of production is limited by ionic diffusion across the membrane. The rate of diffusion across the membrane can be increased by using turbulent flow. The disadvantage of this method is that the pumping power required to circulate the solution in the turbulent flow regime is high, and besides, the degree of conversion per pass may decrease owing to the decrease in the residence time of the solution in the cell. To overcome the disadvantages of using high flow rates in membrane cells, attached and detached turbulence promoters were recommended as a means of enhancing the rate of mass transfer in membrane cells under laminar flow conditions [1–6].

The object of the present work is to explore the possibility of using gas sparging to enhance the rate of mass transfer across the membrane. Gas sparging is gaining increasing importance as a means for enhancing the rate of heat and mass transfer in chemical

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| p | membrane nermeability |
|--------------------|--|
| - | |
| R | cell resistance |
| Re | Reynolds number $(\varrho V_s l/\mu)$ |
| St | Stanton number (K/V_s) |
| Sc | Schmidt number (v/D) |
| ť | time of dialysis |
| $V_{\rm s}$ | superficial gas velocity |
| V | solution volume in each compartment |
| 3 | gas hold-up (void fraction) |
| Q | solution density |
| μ | solution viscosity |
| v | kinematic viscosity |
| θ_0, θ | specific conductivity of electrolyte and |
| | electrolyte-gas dispersion respectively |

and electrochemical reactors in view of its low power consumption compared to mechanical stirring [7–9]. Besides, the combination of gas sparging with low solution flow rates in the continuous operation of membrane separation processes would lead to high degrees of conversion per pass as a result of the high mass transfer coefficient caused by gas sparging and the high residence time of the solution resulting from the low solution flow rate.

2. Experimental technique

Figure 1 shows the dialysis cell used. It was made of Plexiglass and consisted of two similar compartments separated by a membrane. Each compartment was rectangular and had dimensions 5×10 cm for the base and was 12 cm high. Before each run, the two cell halves were assembled with the membrane between them. The membrane was held firmly by the flanges of the two compartments. A high permeability Cuprophane membrane (J. P. Bemberg, Wuppertal, Germany) was used, which had a wet thickness of



Fig. 1. Experimental apparatus.

16.7 μ m and a permeability of 0.001 23 cm s⁻¹. At the bottom of the cell two gas spargers were placed on each side of the membrane.

Preliminary experiments with perforated plastic tube spargers showed that gas distribution was not uniform. Porous ceramic gas spargers were found to give uniform gas distribution at the membrane surface and were used throughout this work. Each distributor had a triangular cross section; the base of the distributor (8 cm \times 1.6 cm) which rests on the cell bottom in the immediate vicinity of the membrane was coated with epoxy to allow gas bubbles to evolve only from the sides of the distributor in the form of a curtain in the immediate vicinity of the membrane. The distributor was blocked at one end and had a circular opening at the other end through which air was fed. The distributor had a porosity of 22%. Since the length of the distributor was 8 cm while the membrane width was 10 cm, the active membrane width was reduced to 8 cm by sticking plastic tape along the edges of the membrane.

Air was driven to the spargers by two variable speed air compressors. Air flow rate was measured by a calibrated rotameter and regulated by controlling the compressor speed. In all experiments the superficial air velocity was adjusted to be the same in each compartment. Superficial air velocity was calculated by dividing the volumetric air flow rate by the projected area of the sparger.

Each run was started by placing 500 cm³ of NaCl in one compartment and 500 cm³ of distilled water in the other compartment. The two compartments were filled with the liquids simultaneously and at the same rate to avoid any difference in solution level between the two compartments. Gas sparging was started immediately after filling the two compartments. The rate of dialysis was followed by measuring the conductivity of the solution with time. A conductivity cell was placed in the concentrated solution bulk, 2.5 cm from the diaphragm. The position of the conductivity cell in the solution was found to have no effect on the conductivity value.

Initial NaCl concentration ranged from 100 to 300 gl^{-1} prepared using AR grade salt and distilled water. Air superficial velocity ranged from 0 to 9.38 cm s^{-1} and the temperature was $25 \pm 1^{\circ}$ C. Preliminary experiments showed that air humidification before entering the cell gave the same results as unhumidified air within the present range of superficial air velocities. So, air humidification was dispensed with. Dialysis was conducted for a maximum time of 60 min. Beyond this time equilibrium between the two compartments was approached especially at high air superficial velocities. The Schmidt number of the solution ranged from 644 to 712.

3. Theory

The overall mass transfer coefficient (K) for the membrane is related to the two liquid phase mass coefficients $(K_1 \text{ and } K_2)$ and the membrane permeability (P) by the equation

$$\frac{1}{K} = \frac{1}{K_1} + \frac{1}{P} + \frac{1}{K_2}$$
(1)

Since a high permeability membrane was used in the present work the overall mass transfer coefficient is mainly determined by the hydrodynamic conditions on the two sides of the membrane. The equation used to calculate K from concentration-time data was derived as follows.

A material balance on the compartment containing concentrated NaCl gives

$$V \frac{dC_1}{dt} = KA (C_1 - C_2)$$
 (2)

For the compartment containing dilute NaCl

$$V\frac{\mathrm{d}C_2}{\mathrm{d}t} = KA\left(C_1 - C_2\right) \tag{3}$$

Combining equations (2) and (3)

$$V \frac{d(C_1 - C_2)}{dt} = -2 KA(C_1 - C_2)$$
 (4)

$$\frac{d(C_1 - C_2)}{(C_1 - C_2)} = -\frac{2KA}{V} dt$$
 (5)

Integrating from t = 0 to t = t gives

$$\ln\left[\frac{(C_1 - C_2)_t}{(C_1 - C_2)_0}\right] = -\frac{2KAt}{V}$$
(6)

Substituting for C_1 by

$$C_1 = C_0 - C_2$$
 (7)

$$\ln\left[\frac{(C_0 - 2C_2)}{C_0}\right] = -\frac{2 KAt}{V}$$
(8)

i.e.

$$\ln\left[\frac{1}{1 - (2C_2/C_0)}\right] = \frac{2KAt}{V}$$
(9)

4. Results

Figure 2 shows that the experimental data fit equation (9) at different air superficial velocities. The overall mass transfer coefficient K was calculated at different air velocities from the slopes of the straight lines shown in Fig. 2. Figure 3 shows the effect of air superficial velocity on the overall mass transfer coefficient. The



Fig. 2. ln $(1/[1 - (2C_2/C_0)])$ against t.

data fit the equation

$$K = aV_{\rm s}^{0.22} \tag{10}$$

The velocity exponent 0.22 is in agreement with the values reported by authors who studied rates of heat transfer in bubble columns. Louisi [10] obtained a value of 0.19, and Steiff *et al.* [11] obtained 0.22, as did Fair *et al.* [12]. Hart [13] obtained a value of 0.25; the theoretical model of Deckwer [14] also gave a value of 0.25.

The velocity exponent also agrees with the values reported by authors who measured rates of mass transfer in gas sparged reactors. Sedahmed *et al.* [15] obtained a value of 0.2 for air sparged vertical screens. Theoretical treatment of mass transfer in gas sparged parallel plate cells [16] led to an exponent of 0.25. Piovano *et al.* [17] and Cavatorta and Bohm [18] correlated their mass transfer data at gas sparged vertical electrodes in terms of the gas hold-up rather than the superficial gas velocity. According to these authors the mass transfer coefficient can be related to the gas hold-up for vertical electrodes by the equation

$$K \propto \varepsilon^{0.246}$$
 (11)

The hold-up data of Cavatorta and Bohm [18] show that below the plateau the gas hold-up can be related to superficial velocity with fair accuracy by the equation

$$\varepsilon \propto V_{\rm s}.$$
 (12)

Accordingly, the data of Piovano *et al.* and Cavatorta and Bohm can be related by the equation

$$K = a_1 V_s^{0.246}. (13)$$

The exponent 0.246 agrees fairly with the present value. The present $\log K/\log V_s$ slope is at variance with the values 0.34 and 0.33 reported for the case of heat transfer in bubble columns [14]. Also the present $\log K/\log V_s$ slope differs from the value 0.378 obtained for mass transfer at gas sparged horizontal single screen electrodes [19] and the value 0.36 obtained from the mass transfer data obtained by Ibl *et al.*, [7] for gas sparged vertical electrodes. The mass transfer data of Piovano *et al.* [17] for gas sparged vertical expanded metal electrodes can be represented by the equation

$$K = a_2 V_s^{0.311} \tag{14}$$

which gives a $\log K/\log V_s$ slope higher than the present value.

5. Discussion

The discrepancy in the value of the log $K/\log V_s$ slope among different authors can be explained by the assumption that mass transfer in gas sparged cells takes place through a surface renewal mechanism, as explained earlier by Sedahmed [20]. According to this model the mass transfer coefficient can be related to the gas hold-up by the equation

$$K = a_3 \varepsilon^{0.33}. \tag{15}$$

The gas hold-up (ε) is usually related to V_s by Equation (18):

$$\varepsilon = bV_s^n \tag{16}$$

The value of *n* was reported by Shah *et al.* [21] to lie between 0.7 and 1.2 in the bubbly regime. Combining Equations (12) and (13) shows that the log $K/\log V_s$ slope should range from 0.23 to 0.4, which agrees with previous studies in heat and mass transfer. Although gas hold-up depends mainly on superficial velocity it seems that this dependence is very sensitive to the physical properties of the solution and the presence of



Fig. 3. Effect of superficial velocity on the overall mass transfer coefficient.



trace impurities [21]. This may explain the large scatter in the hold-up data reported in the literature and explain the difficulty of obtaining a unified correlation between the gas hold-up and the superficial velocity [21]. According to the surface renewal model [20], the discrepancy in the gas hold-up data may explain the difference in the value of $\log K/\log V_s$ slope obtained by different authors. Besides, the difference in the value of the log $K/\log V_s$ slope obtained by Bohm et al. [17, 18] for vertical plate and vertical expanded metal electrodes, as shown by Equations (16) and (17), suggests that the turbulence-promoting ability of the transfer surface plays a role in determining the $\log K/\log V_s$ slope. Equations (13) and (14) show that the log $K/\log V_s$ for a vertical expanded metal electrode is higher than that for the vertical plate electrode under otherwise indentical conditions by virtue of the ability of an expanded metal electrode to promote turbulence. Also the fact that the $\log K/\log V_s$ slope for gas sparged horizontal single screens [19] is higher than that of vertical single screens [15] substantiates the role played by the geometry of the transfer surface in determining the log $K/\log V_s$ slope.

6. Data correlation

To throw some light on the effect of other variables, such as the physical properties of the solution, an overall mass transfer correlation was envisaged in terms of the dimensionless groups J, Fr and Re usually used to correlate heat transfer data in bubble columns [14]. Figure 4 shows that the data fit the equation

$$J = 0.062 \, (Fr \, Re)^{-0.26} \tag{17}$$

with an average deviation of $\pm 15\%$. The physical properties (ϱ , μ , D) used in calculating the dimensionless groups were taken as the mean of the time-averaged values for the two compartments. The physical properties at different NaCl concentrations were taken from the literature [22]. In the area of heat transfer Hart [13] correlated heat transfer rates in bubble agitated systems by the equation

$$J = 0.125 \, (Fr \, Re)^{-0.25}. \tag{18}$$



Mass transfer data at vertical gas sparged single screen electrodes [15] were correlated by the equation

$$J = 0.187 \, (Fr \, Re)^{-0.26}. \tag{19}$$

It should be emphasized that the coefficient 0.062 of equation (17) cannot be applied to other membranes because it depends, in part, on the membrane permeability which is a function of the membrane structure and the nature of the diffusing substance [23].

7. Practical applications

In view of the substantial increase in the rate of mass transfer at the membrane surface, which ranged under the present conditions from 18 to 360%, over the natural convection value, gas sparging can be used in electrodialysis stacks to eliminate concentration polarization and reduce the diffusion layer thickness at the ion exchange membrane surface with a consequent increase in the limiting current and the rate of desalination. The limitation of using gas sparging in electrodialysis is that the presence of a bubble curtain on the two sides of the membrane may increase the IR drop and power consumption [24]. Consider for example the case of gas hold-up (ε) = 10%, the conductivity of the solution [25] will be $\theta = \theta_0 (1 - \varepsilon)^{1.5} = 0.85 \ \theta_0$ and the power consumption will increase by about 17%. From the hold-up data of Cavatorta and Bohm [18] the superficial velocity which corresponds to $\varepsilon = 10\%$ ranges from 2-4 cm s⁻¹. For this range of gas velocity the present results show that the mass transfer coefficient increases by an amount ranging from about 50 to 100%, depending on NaCl concentration. Since electrodialysis is conducted in practice using flowing solutions, it is expected that the adverse effect of gas sparging on cell voltage will decrease [26]. However before using gas sparging in practical electrodialysis a pilot plant study is needed to optimize the operating conditions. Gas sparging can be used with greater advantage in other non-electrical membrane separation processes where the above limitation is nonexistent, such as dialysis, hyperfiltration and reverse osmosis, to eliminate concentration polarization and increase the rate of separation.

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